

Serial No. 09/247,219 - Peggy M. Tomasula

Applicant wishes to thank the Examiner for the detailed explanation of the rejection under 35 U.S.C. §103(a). The rejection and the Examiner's Response to Applicant's arguments are discussed below.

The Rejection under 35 U.S.C. §103(a).

Claims 2, 4-6, 8-9, 11-13 and 15-16 were rejected under 35 U.S.C. §103(a) as being unpatentable over Dahlstrom et al. taken with Tomasula '265. Withdrawal of this rejection is requested for the following reasons. Applicant's remarks submitted in the previous responses are incorporated herein.

The Applicant fully recognizes that the rejection was based upon the combination of Dahlstrom in view of Tomasula. Any discussion in Applicant's remarks of either reference individually was for the Examiner's benefit in understanding what each reference teaches alone, so that the existence of motivation to combine the teachings of the respective references could be properly assessed. The point the Applicant wished to make in the previous response was that there could not have been sufficient motivation to combine Dahlstrom and Tomasula. If Dahlstrom were modified to accommodate the gaseous carbon dioxide of Tomasula, then Dahlstrom would become inoperative and its intended function

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would be destroyed. (See *In re Fritch*, 972 F.2d 1260, 1265-66, 23 U.S.P.Q. 2d 1780 (Fed. Cir. 1992) ("A proposed modification [is] inappropriate for an obviousness inquiry when the modification render[s] the prior art reference inoperable for its intended purpose.")). Applicant apparently was not convincing in this regard, and submits the following information for the Examiner's consideration.

Dahlstrom requires treatment of a protein-containing stream and a reactant stream for coagulating the protein in a resonating chamber at ultrahigh frequencies (see Summary in col. 2, para. bridging cols. 2 and 3, and Claim 1). The reactant stream is preferably a food grade acid, such as carbonic acid (col. 3, lines 49-65). A commercial embodiment of a device which provides the requisite ultrahigh frequencies in a resonating chamber is the Sonolator™ produced by Sonic Engineering Corporation of Stratford, Connecticut (col. 3, lines 13-22). This is the only commercial embodiment disclosed by Dahlstrom, and may in fact be the only device available on the market having the requisite features for Dahlstrom's applications.

What is a Sonolator™? Sonic Corporation's Web Site (5 pages submitted herewith) defines it as an ultrasonic homogenizing device that employs high pressure and cavitational forces. It

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operates by passing one or more **liquid** streams at high velocity over a blade. The blade acts as a foil to create a field of extreme cavitation. The material is forced into the Sonolator™ by means of a positive displacement pump. It is important to note that the material must be in a liquid stream. Submitted herewith are excerpts from the Berliner Web Site regarding ultrasonics. On the page headed "Ultrasonics and Fine Particles" it clearly states, "Cavitation can not occur in air, gas or vapor." On the page headed "Ultrasonic Cavitation", it states:

"Ultrasonic Processing" means "blasting" liquids, usually water, with very intense sound at high frequency, producing very good mixing and powerful chemical and physical reactions. This process, called "cavitation", is sort of "cold boiling" and results from the creation and collapse of zillions of microscopic bubbles in liquid."

The ensuing discussion in the aforementioned Berliner reference describes why the process of cavitation requires a liquid medium.

Suslick (submitted herewith) discusses the chemistry of ultrasound. On the first page, paragraph 3, Suslick states that "cavitation occurs in liquids not only during turbulent flow but also under high-intensity ultrasonic irradiation." At the top of the next page, Suslick unequivocally states, "Because cavitation can take place only in liquids, chemical reactions do not generally occur during the ultrasonic irradiation of solids or **solid-gas** systems." (emphasis added).

Applicant submits that a person of ordinary skill in the art would immediately recognize from the information discussed above that it would be impossible to substitute gaseous carbon dioxide for the liquid carbonic acid of Dahlstrom and retain the functionality of the Dahlstrom process. Introducing a gaseous stream into a Sonolator™ or any other ultrasonic device having a resonating chamber as required by the reference would render such device inoperable. No further analysis is necessary to defeat the rejection. However, each of the points raised by the Examiner is addressed below.

Step (a) of Claim 15 requires that the starting material has a protein concentration of less than 80% by total weight of solids and step (e) requires the recovered protein precipitate has a concentration of protein greater than 85% by total weight of solids. The Examiner merely speculates that the protein source of Dahlstrom would have less than 80% protein, though there is no indication in the reference that this is indeed the case. The Examiner's statement that Applicant's claim is open-ended and therefore does not exclude other sources of protein is not fully understood. The fact that other sources of protein might be included within the scope of the claims does not speak to the protein concentration. For example, three sources of

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protein, each having a 70% concentration, would provide a composite also having a protein concentration of 70%.

The Examiner also intimates that the "comprising" language is permissive of other levels of protein. The "comprising" language in the preamble applies to the ensuing steps. The protein concentrations in steps (a) and (e) are positive recitations that serve as limitations to the scope of the claims. These limitations cannot be ignored, just as the process steps themselves cannot be ignored.

The teaching in Tomasula of using carbon dioxide to precipitate milk proteins is not disputed. And it is true that some of the carbon dioxide will dissolve in the aqueous medium. However, that is not sufficient basis for using carbon dioxide and carbonic acid interchangeably for the precipitation of vegetable proteins as suggested by the Examiner. Though the reduction in pH caused by dissolution of the carbon dioxide can contribute to the precipitation of casein from milk, that is not to say that dissolved CO₂ would have the same effect on vegetable proteins; or that the pressurized gaseous CO₂ does not play a role in the properties of the precipitated product. Assuming *arguendo* that a fluid stream of gaseous carbon dioxide could be used in ultrasonic device of Dahlstrom (which it cannot, as shown

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above), the extreme cavitation would reduce the vegetable curd to very small particles sizes (below 0.10 microns) as reported on the first page of the Sonic Corporation's Web Site reference discussed above. In contrast, Applicant does not use cavitational forces, but rather holds vegetable protein in solution/dispersion for at least one minute under a headspace of carbon dioxide (page 6, lines 15-16). When the pressure is gradually released as required by step (c) of Claim 15, the particle size of the protein precipitate is maintained (see page 7, lines 16-19, of specification). The Examiner's point regarding Tomasula's use of a high pressure pump is not understood. Regardless of the apparatus used, it is significant that Tomasula **gradually** reduces the pressure and does not subject the precipitate to forces that will substantially atomize it.

At the bottom of page 4 of the Office Action, the Examiner states that the source of the pressure is not disclosed in the claimed invention. The source of the pressure on the solution/dispersion **is** the carbon dioxide. If the Examiner's hand were placed under a steel press bearing down at 800 psi (no ill will intended, just an analogy), certainly the Examiner would agree that it was the steel press applying the pressure. It is the same with the CO₂ bearing down on the surface of the

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vegetable protein dispersion/solution. Applicant does not desire to be limited to any particular apparatus for introducing the CO₂ to the treatment chamber and are not aware of any requirement to do so, especially in a process claim.

Regarding the Examiner's discussion of the holding step (top of page of Office Action), Applicant acknowledges that Tomasula discloses a holding step. That is well and good; but for the rejection to stand, there must be a reason why a person of ordinary skill in the art would be inclined to modify the primary reference to include a holding step taught in the secondary reference. The ultrasonic treatment contemplated by Dahlstrom simply does not allow for a holding step. The protein-containing fluid and the acid (whether as separate or a combined stream) are blasted through the resonating chamber like water through a fire hose. The residence time in the device is disclosed as being less than 0.628 seconds (col. 4, lines 37-44). There is no provision (or reason) for holding the protein stream for at least 1 minute as required by step (b) of Claim 15.

The Examiner also urges that Tomasula discloses a separation step. Just as with the holding step, there is no provision in Dahlstrom for separating the protein from the acid coagulant. In fact, Dahlstrom does just the opposite. The high shear forces of

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the ultrasonic treatment uniformly disperses the ingredients of the two feed streams. The dispersed protein and acid react to yield an acidified protein reaction product (col. 4, lines 13-36). Following this reaction, there is nothing left to separate. In contrast, when the Applicant's solution/dispersion is depressurized, the carbon dioxide and carbonic acid vaporize (page 3, lines 18-19, of specification) and are thereby separated from the vegetable protein. The recovered product has a pH approximating the original pH of the dispersion/solution before introduction of the CO₂ (see p. 8, lines 20-22, of the specification). Applicant urges that the claimed invention is unobvious because it recovers a product having nearly unaltered pH and a cottage cheese curd-like appearance (page 8, lines 24-26) and avoids the formation of contaminating salts (page 8, lines 2-23). There is nothing in either Dahlstrom or Tomasula to indicate that vegetable protein could be rendered into a curdlike product. The significance of the concentration of the protein in the recovered product being greater than the concentration of the protein in the starting material is to emphasize that there is indeed a physical separation of a protein curd from other non-curd components, unlike what happens in Dahlstrom.

It is unclear where the Examiner is going with the statements about the order of the processing steps (paragraph bridging pages 6 and 7 of the Office Action). However, the language of Claim 15 does not permit performing the various recited steps in any random order. Step (a) is drawn to **pressuring** the vegetable protein dispersion/solution. Step (b) **holds** the pressurized dispersion/solution [from step (a)] at a specific pressure and for a specific time to form a precipitate. Step (c) gradually **depressurizes** the dispersion/solution in order to maintain the particle size precipitate formed in step (b). Obviously, this must take place after the holding in order for the holding step to have any significance. Step (d) **separates** said protein precipitate (namely, that described in steps (b) and (c)). Finally, step (e) recovers a solid protein precipitate. To alter the order of these steps would require distorting the inherent sequence of steps that must be followed.

Applicant respectfully submits that the Examiner's citation of *In re Levin* is misplaced. Applicant's claimed process is **not** merely a recipe or a formula for cooking food. It is a method of processing a component (protein) of a raw material (vegetable matter) in a novel and unique manner in order to obtain a product having specific physical properties. Applicant has demonstrated,

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above, that the vegetable protein and carbon dioxide indeed coact with one another to yield a new and unexpected product having a useful function as, for example, a food ingredient.

Summary.

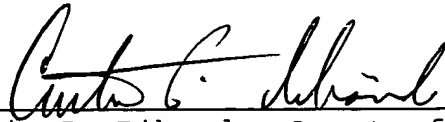
Applicant has delineated in detail the differences between the claimed invention and the teachings of the references. To reiterate, the claimed invention differs from the process of Dahlstrom in several critical respects: (1) Applicant uses high pressure **carbon dioxide** rather than carbonic acid. (2) Applicant applies a **holding step** at a pressure in the range of 400-800 psi which Dahlstrom does not do. (3) Applicant employs a **gradual** depressurization in step (c) which Dahlstrom does not do. (4) Applicant **separates** a protein precipitate from the initial vegetable protein source, which Dahlstrom does not do. (5) Applicant **concentrates** the protein from an initial 80% by total weight solids to at least 85% by total weight solids. The instantaneous, ultrasonic treatment of a liquid stream in Dahlstrom process of Dahlstrom does not permit any of these conditions to occur, and cannot be modified to accommodate a gaseous stream as taught in Tomasula.

Accordingly, Claims 2, 4-6, 9, 11-13, and 15-19

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are deemed to be in condition for allowance and a favorable action on the merits of the case is earnestly solicited.

Respectfully submitted,



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Enclosure

1. Sonic Corporation Web Site (5 pgs.)
2. Berliner Web Site (4 pgs.)
3. Kenneth S. Suslick, "The Chemistry of Ultrasound" (2 pgs.)

Sonolator What the *HECK* is a Sonolator?

High-Pressure Ultrasonic Mixing and Homogenizing Systems

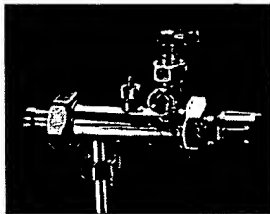
The Sonolator is an in-line, High-pressure Ultrasonic Homogenizing device. We employ high pressure and cavitation forces to create fine emulsions and dispersions ideal for use in a wide range of applications. As seen here, our systems force material through a specially engineered orifice, subjecting material to various forces of pressure. Material at high velocity flows over a blade which acts as a foil to create a field of extreme cavitation. The pressure and cavitation work in unison to create very small particle sizes, below 0.10 micron, and very tight size distributions.



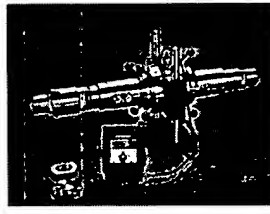
Sonolator by itself in various models:



Model A; 0.5 - 5 GPM,
5000 PSI



Model BT; 5 - 20 GPM,
5000 PSI



Model XS; 20 - 150
GPM, 5000 PSI

Sonolator System Advantages

Extremely Efficient - Processes emulsions and dispersions at as much as **50% the pressure** required by conventional high-pressure homogenizing machines

Time Saver - Processes emulsions and dispersions in a single pass, reducing processing times **as much as 50%** in some cases.

Multiple Feed Systems - Several phases can be metered into the Sonolator via PD pumps to create an instant emulsion; this leads to savings in batch times, elimination of large mixing vessel, compounding times, etc. Continue below to learn more about Multiple Feed Sonolator Systems.

Cost Savings - Cost savings are realized by energy reductions and time savings gained by elimination of recycle times in single pass operation, elimination of large intermediary mixing vessel and reduced operating pressure which mean less motor horsepower and less amperage draw.

Click Here...

To learn about Standard Sonolator Homogenizing System Features

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Standard High-pressure Ultrasonic Sonolator Homogenizing Systems

As mentioned previously, the **Sonolator** is incorporated into comprehensive homogenizing systems. The Sonolator is the focal point of every system where all emulsification, dispersion and mixing takes place. A positive displacement pump is required to force material into the Sonolator. In our line of Standard Pilot Sonolator Systems, a triplex plunger or piston pump is incorporated, allowing flow rates to 5 GPM and pressures to 5000 PSI. These systems feature:

- Premium efficiency motors by US Motors, Baldor, Siemens, Reliance and more
- Pressure gauge on a 316 stainless steel seal assembly
- Full-flow, 316 stainless steel pressure relief device
- Premium quality Variable Frequency Drives by ABB, Allen Bradley, Siemens, Zener and more
- An operator station including system speed adjusting knob and start/stop buttons.
- A stainless steel tubular base on casters with brake or on platform feet

In the Standard System the customer can choose which motor and drive brand they desire and the input power requirements. You as a customer can also choose to include these accessories:

- Electronic pressure transmitter with digital display and 4-20 mA output connection to PLC.
- Electronic pressure switch with 4-20 mA output connection to PLC to terminate system if dangerously high pressures are encountered.
- Flow meters
- Flow switches
- Digital tachometer
- Electronic temperature probe mounted in a stainless steel thermocouple well
- Heat exchangers

[Click Here...](#)

To see our Standard Benchtop and Pilot Application Sonolators

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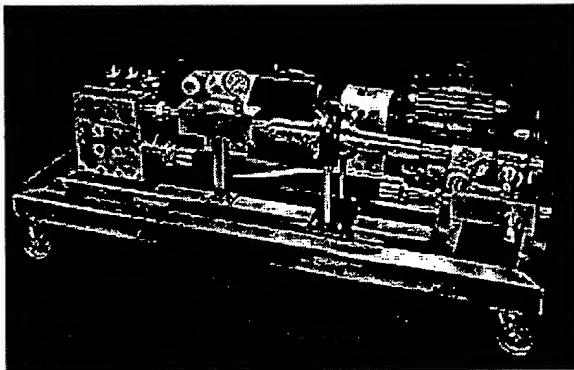
Custom Designed Sonolator Systems

In addition to the Standard Sonolator Systems which employ the triplex piston and plunger pumps, Sonic fabricates custom designed systems to meet various customer needs including systems. As previously mentioned, we can furnish any style PD pump required given your application. We have several Sonolator Systems in the field featuring:

- Progressing cavity pumps
- Gear pumps
- Diaphragm pumps
- Lobe pumps and more!
- Pressures to 1500psi
- Flow rates to 150 gpm
- Multiple Feed Scenarios

The ability to use so many different style pumps gives Sonic the opportunity to provide you with a wealth of options when designing your homogenizing system. The Sonolator, having no moving parts, shows no discretion to viscosity; as long the right pump has been selected to force the material into the device, homogenization of materials as thick as peanut butter will be achieved. Sonic works closely with you, the customer, to design the layout and select proper, well known and trusted components and instrumentation. Layout drawings, flow diagrams, electrical schematics are drawn on CAD at Sonic by trained engineers. Our custom designs can include:

- Flow Meters and switches
- Tanks and vessels
- Pressure transmitters and switches
- Mixers and agitators
- Complete PLC integration and programming
- Heat exchangers
- Control valves
- Variable frequency drives and enclosures
- Sonic works with you to make sure the design meets all your processing requirements.



Production SonolatorTM 2000-30

30 gpm, 2000 psi. All results obtained on the Benchtop or Pilot SonolatorTM Systems can be scaled up directly and duplicated on Production SonolatorTM Systems. This unit features a triplex plunger pump with block-style manifold, a 40 horsepower motor and a Sonic XS1500 SonolatorTM. This system can be used for a wide range of applications including hand creams, lotions, chemical emulsions, tomato sauces and juices. Production times are cut by nearly 20 to 30% when this system is used in place of batch mixers.

CONTINUE... to learn more about ***Multiple Feed Sonolator Homogenizing Systems***

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Multiple-Feed Ultrasonic Sonolator™ Homogenizing Systems

As mentioned previously, the Sonolator has the ability to process a number of liquid streams to instantaneously create a finished emulsion or mixture in a single pass. Several PD pumps are installed into the system which accurately meter various phases at specified ratios into the Sonolator. Giving you the option to meter in different phases provides several benefits:

Reduced Process Time and Costs: Time consuming batch processes requiring excessive compounding and heating can be eliminated.

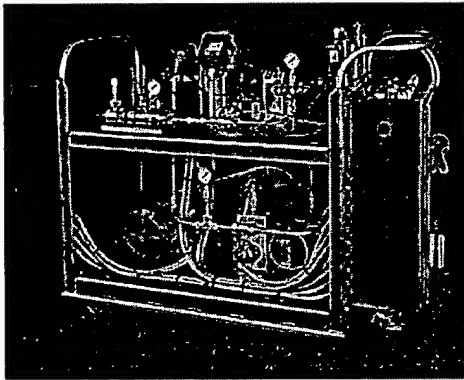
Heating Reductions: With individual phases, you need heat only those phases requiring it, such as oils and waxes.

Tank Reduction: Smaller, less expensive and more manageable tanks can be used in the Multiple-Feed approach.

Avoids Viscosity Increase Problems: The Multiple-Feed approach is very useful in processing materials where viscosity increases dramatically upon mixing.

Product Tuning: Various renditions of the same product can be made simply by altering feed ratios.

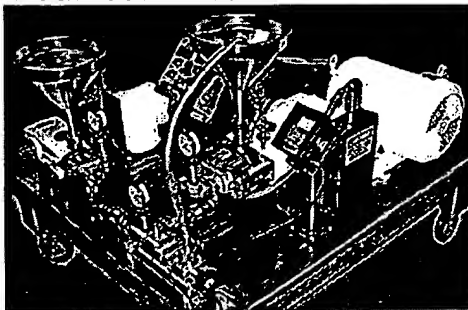
Dual-feed Production Sonolator 1200; 15 GPM, 1200 PSI



This system is being used by a Textile Fiber producing company to process spin finish emulsions. The system is fully automated by controls designed and installed by Sonic. The system is portable and can process a broad range of emulsions on

demand at various feed ratios. Previously, the customer spent several hours processing in large mixing vessels. This system makes material on demand, eliminating the large, intermediary mixing vessel and reducing production times by as much as 20%.

Dual-feed Pilot Sonolator 2000; 6 GPM, 2000 PSI



This system is being used by a Personal Care Products company to process creams and lotions. The oil and water phases are metered in separately to form a uniform, stable

emulsion instantaneously. Additional feeds are added for aloes, fragrances, etc. With this approach, costly mixing vessels can be eliminated; only those phases requiring it need be heated, other phases can be metered in at ambient temperatures; greater control is gained over processing; compounding errors are reduced; and waste and wax/oil consumption can be

reduced by as much as 20%. The system incorporates manual controls designed and installed by Sonic. The system can process a wide range of emulsions on demand at various feed ratios.

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ULTRASONICS AND FINE PARTICLES - BENEFICIATION OF SLURRIES AND FINE-PARTICLE SUSPENSIONS [CERAMICS, COAL & ORES, COATINGS, COLUMN PACKINGS, SINTERING, SLIPS]

AL-2

ULTRASONICS AND FINE PARTICLES -

Jan 98

BENEFICIATION OF SLURRIES AND FINE-PARTICLE SUSPENSIONS

[CERAMICS, COAL & ORES, COATINGS, COLUMN PACKINGS, SINTERING, SLIPS]

(Also including suggestions for best mixing of batch samples -
"sifting solids and swirling solutions")

I. GENERAL -

Sonication of suspensions of ultrafine particles provides a number of significant benefits, not the least of which is better dispersion. Ultrasonics substantially reduces particle size of ultrafine suspensions in one tenth the time of traditional ball milling methods. In addition, one can expect disaggregation and deagglomeration of clumps (particle size reduction), degassing of the carrier liquid, increased slurry flow properties, higher homogeneity, and denser castings, sinterings, or packings.

Cavitation, the formation and implosion of microbubbles in a high-intensity ultrasonic field, propagates shock waves through the liquid. This intense energy accelerates both physical and chemical reactions, enhancing surface chemistry and causing violent particle motion and generating high-velocity interparticle collisions.

Bubble formation occurs in the liquid between particles. Cavitation can not occur in air, gas, or vapor. Thus, no action is found in unwetted, gas-filled voids in a particulate mass. For an insoluble material suspended in an inert liquid, the effective viscosity of the parent liquor (that property of the suspending liquid affecting cavitation) is just that of the basic liquid, which can be quite low, and not the apparent viscosity of the suspension, which can be quite high. For this reason, it is possible to sonicate extremely dense suspensions or slurries in water or light solvents. Such thick slurries might have apparent viscosities far above the range of 5,000 to 10,000 centipoise (5 to 10 Pa.s), which is the threshold of cavitation for most simple liquids.

Wetted beds of particles can be fluidized with probes or in cup horns or even in ultrasonic cleaners for laboratory-scale experimentation.

Field experience has borne out these ideas. Several practical examples follow:

a. Ceramic insulation for resistors and capacitors benefits greatly from the homogeneity and degassing provided by sonication. Probes are used in conjunction with continuous flow cells to give on-line production capability in a number of electronics applications.

b. Iron oxides and similar disk and tape coatings are dispersed and degassed immediately prior to

application in slurry form. Freedom from voids and smears provides a far superior product for manufacturers of magnetic media.

c. Clays, fines, and other fines can be compacted (dewatered) ultrasonically. Significant development work has been done in this area. Compacting of soil samples and selectively increasing or decreasing permeability are also practicable. In addition, a phenomenon of gelling of clays, especially in the presence of petroleum-based oils, has been noted by the author.

d. Coal beneficiation and ore refining obtain greater yields and coal slurries can be made denser for better transport and improved combustion properties.

e. Pharmaceutical preparations and tablet pressing operations are also areas in which sonication of fine particle suspensions has been applied. Reduction of tablet size is a very attractive application. [Tablet producers should note that ultrasonic liquid processors are also used for dissolution of samples for faster QC analysis.]

f. Glass beads (microspheres) are commonly used as column packings for filtration and for HPLC (High Pressure Liquid Chromatography) and similar applications. Sonication of the suspended beads immediately prior to packing the column has repeatedly resulted in 20% denser packing. This allows 20% shorter equivalent columns or a 20% higher performance in the same column length.

g. Industrial ceramics and even fine china table service can be improved by sonicating the slip prior to pouring the mold. Better slip homogeneity, fewer voids from bubbles, smoother surface finishes, and less cold jointing result. Probes can be fitted directly to slip pouring nozzles at molding stations.

h. Sintered carbide tool bits are made by suspending fine particles of tungsten or other carbides in light fluorocarbon liquid, pouring the suspension in a die cavity, evaporating (and recovering) the solvent, and pressing the particles in the cavity under extreme pressure and temperature. By sonicating the particles either immediately before pouring, or even directly in the cavity, a 20% improvement in density was found. Resultant bits are stronger, hold an edge longer, cut cleaner, and remain cooler. Thinner bits can be made, thus providing cost savings.

i. Photographic emulsion grains can be sonicated prior to coating to degas and homogenize the material and to prevent formation of voids and discontinuities.

In addition, ultrasonic processing of suspensions in chemically-reactive liquids provides greater yields through acceleration of surface chemistry. A new field, sonochemistry, has, in part, resulted from the realization of the ability of cavitation to both expose fresh surface and enhance reactions.

2. SIFTING SOLIDS AND SWIRLING SOLUTIONS

The ability of an ultrasonic liquid processor to effectively stir, mix, or agitate a batch depends to a large degree on the sample volume being appropriate for the horn and tip being used. Such sample volumes are usually indicated in manufacturers' catalogs. On occasion, however, it becomes necessary to process volumes larger than recommended. While inefficient, wasteful of tips, and time consuming, the procedure can be improved if appropriate steps are taken:

a. In any batch sonication procedure, the shape and size of the vessel can be critical. In general, beakershaped vessels are best, and round-bottomed vessels such as test tubes and boiling flasks are fair, while square-bottomed vessels or complex shapes are poor. Vessels with no free space around the tip

Professor of Mechanical Engineering and Director of the Acoustics Research Center at The Cooper Union. Dr. Raichel is a Fellow of the Acoustical Society of America and of the American Society of Mechanical Engineers and also works at CUNY - the City University of New York. This book will focus on the practical application of power (high intensity) ultrasonics, the use of ultrasonic energy to change materials. Contributions are welcome (see below).

THE CAVITATION BUBBLE



[image from University of Washington, Applied Physics Laboratory (Lawrence Crum, Ph.D.)
- bubble diameter approximately 1mm]

ULTRASONICS - continued

ULTRASONIC CAVITATION

[See the photo of a cavitation bubble at the top of this page
and the section, More on Cavitation, on page 2]

AL-1V

A POPULARIZED GUIDE TO ULTRASONIC CAVITATION

4-97

(A Non-Technical Explanation of "Cold Boiling")

"Ultrasonic Processing" means "blasting" liquids, usually water, with very intense sound at high frequency, producing very good mixing and powerful chemical and physical reactions. The process, called "cavitation", is sort of "cold boiling" and results from the creation and collapse of zillions of microscopic bubbles in the liquid.

"Cavitation" or "cold boiling" is easy to understand if you think about what the words "solid", "liquid", and "gas" mean.

A solid is something hard that you can see and touch and hold; its molecules can not move in relation to each other; they are "stuck together".

A liquid is something you can see and touch, but it runs through your fingers if you try to hold it without a cup or a bowl; its molecules are free to move around each other but they can't move apart. That means that they are "slippery"; they can flow.

A gas is something you can touch, like the wind moving across your hand when you stick it out the window of a moving car, but you can't usually see it and you can't hold it at all without a closed can or bottle; its molecules are free to move around and together or apart from each other. They can expand or contract without limit.

The definition in physics of a solid is something whose molecules are rigidly bound together in time and space, a liquid is something whose molecules are free to move around each other at a fixed distance, and a gas is something whose molecules are free to move around each other and to move closer together or further apart.

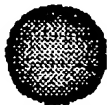
You know you can bend a solid, like bending a branch or matchstick or toothpick. If you bend it too far, it snaps. If you bend a paper clip back and forth enough times, you can break it, too; you "fatigue" the metal or wear out the bond that holds the molecules together. What you are doing in each case is called "exceeding the elastic limit"; you are bending it further than it can bend without breaking. With a hammer, you can break a brick or a small stone. With a big enough hammer or a wrecking ball, you can smash rock or boulders or concrete.

Well, you can break liquids, too! You do it every time you break glass! Glass isn't really a true solid; it is actually a very, VERY, VERY thick liquid, sort of like a super thick syrup or molasses. If you look carefully at ancient window glass, you can see that it has drooped; it has a bulge toward the bottom of the pane. That's because it is flowing downhill; gravity is pulling it down even though it's held in the window frame. "Silly Putty" is exactly the same thing, only not quite as thick; you can see it flow if you wait long enough. But hit it or snap it and it breaks.

Just as you broke the paper clip by bending it back and forth slowly, you can break water (or most other liquids) by jiggling it back and forth, only you have to do it very quickly. By sticking a vibrating object into water, if you vibrate it far enough (a tiny fraction of an inch) and fast enough (around 10,000 times a second), you can "fatigue" the water and break the bond between the water molecules. But what does that mean? What was the definition of a gas? Something whose molecules could move apart. So, if you move water molecules apart, you have a gas, and the gas of water is steam. A steam bubble is normally created by heating above the boiling point (212°F or 100°C). But we just did it by fast jiggling, not by heating, so we "cold boiled" the water!

Next, we now have a steam bubble wandering around in a cold liquid, and that just can't be! The steam has to condense (the way steam from a kettle or hot shower frosts a glass or mirror) and that leaves an empty space behind, a "void" or "cavity", where the steam was. The surrounding water molecules rush in to fill that cavity; when they reach the center of the cavity, they collide with each other with great force. This is called "cavitation". That makes the molecules bounce back, creating a "shock wave" which runs outward from the collapsed bubble just like ripples in a pond when you throw in a pebble. The shock wave can wear away metal; like the edges of an outboard motor propellor. Cavitation was discovered by investigating why propellers wear out.

Where shock waves meet each other, they can cause more steam bubbles to occur and collapse, creating



THE CHEMISTRY OF ULTRASOUND

by Kenneth S. Suslick

from *The Yearbook of Science & the Future 1994*;
Encyclopaedia Britannica: Chicago, 1994; pp 138-155.

Overview

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Ultrasound can produce temperatures as high as those on the surface of the Sun and pressures as great as those at the bottom of the ocean. In some cases, it can also increase chemical reactivities by nearly a millionfold.

Ultrasound is simply sound pitched above human hearing. It has found many uses in many areas. At home, we use ultrasound for dog whistles, burglar alarms, and jewelry cleaners. In hospitals, doctors use ultrasound to remove kidney stones without surgery, to treat cartilage injuries (such as "tennis elbow"), and to image fetal development during pregnancy. In industry, ultrasound is important for emulsifying cosmetics and foods, welding plastics, cutting alloys, and large-scale cleaning. None of these applications, however, take advantage of the effects that ultrasound can have on chemical reactivity.

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The chemical applications of ultrasound, "sonochemistry", has become an exciting new field of research during the past decade. The history of sonochemistry, however, begins in the late 1800s. During field tests of the first high-speed torpedo boats in 1894, Sir John I. Thornycroft and Sydney W. Denaby discovered intense vibrations from and rapid erosion of the ship's propeller. They observed the formation of large bubbles (or cavities) formed on the spinning propeller and postulated that the formation and collapse of these bubbles were the source of their problems. By increasing the propeller size and reducing its rate of rotation, they could minimize this difficulty of "cavitation". As ship speeds increased, however, this became a serious concern and the Royal Navy commissioned Lord Rayleigh to investigate. He confirmed that the effects were due to the enormous turbulence, heat, and pressure produced when cavitation bubbles imploded on the propeller surface. In the same work, he explained that cavitation was also the origin of teakettle noise!

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This phenomenon of cavitation occurs in liquids not only during turbulent flow but also under high-intensity ultrasonic irradiation. It is responsible for both propeller erosion and for the chemical consequences of ultrasound. Alfred L. Loomis noticed the first chemical effects of ultrasound in 1927, but the field of sonochemistry lay fallow for nearly 60 years. The renaissance of sonochemistry occurred in the 1980's, soon after the advent of inexpensive and reliable laboratory generators of high-intensity ultrasound.

Scientists now know that the chemical effects of ultrasound are diverse and include substantial improvements in both stoichiometric and catalytic chemical reactions. In some cases, ultrasonic irradiation can increase reactivities by nearly a millionfold. The chemical effects of ultrasound fall into three areas: homogeneous sonochemistry of liquids, heterogeneous

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sonochemistry of liquid-liquid or liquid-solid systems, and sonocatalysis (which overlaps the first two). Because cavitation can take place only in liquids, chemical reactions do not generally occur during the ultrasonic irradiation of solids or solid-gas systems.

Frontpiece. This micrograph shows interparticle collisions induced by ultrasound between tin and iron particles about 20 microns in size. The velocity of such collisions can be as high as 500 m/s (1100 mph). The elemental composition dot map was produced by scanning Auger electron spectroscopy and show tin in orange and iron in blue.

Ultrasonic irradiation differs from traditional energy sources (such as heat, light, or ionizing radiation) in duration, pressure, and energy per molecule (Figure 1). Because of the immense temperatures and pressures and the extraordinary heating and cooling rates generated by cavitation bubble collapse, ultrasound provides an unusual mechanism for generating high-energy chemistry. As in photochemistry, very large amounts of energy are introduced in a short period of time, but it is thermal rather than electronic excitation. High thermal temperatures are reached. Furthermore, sonochemistry has a high-pressure component, which suggests that it might be possible to produce on a microscopic scale the same large-scale conditions produced during explosions or by shock waves (a shock wave is a compressional wave formed whenever the speed of a body or fluid relative to a medium exceeds that at which the medium can transmit sound).

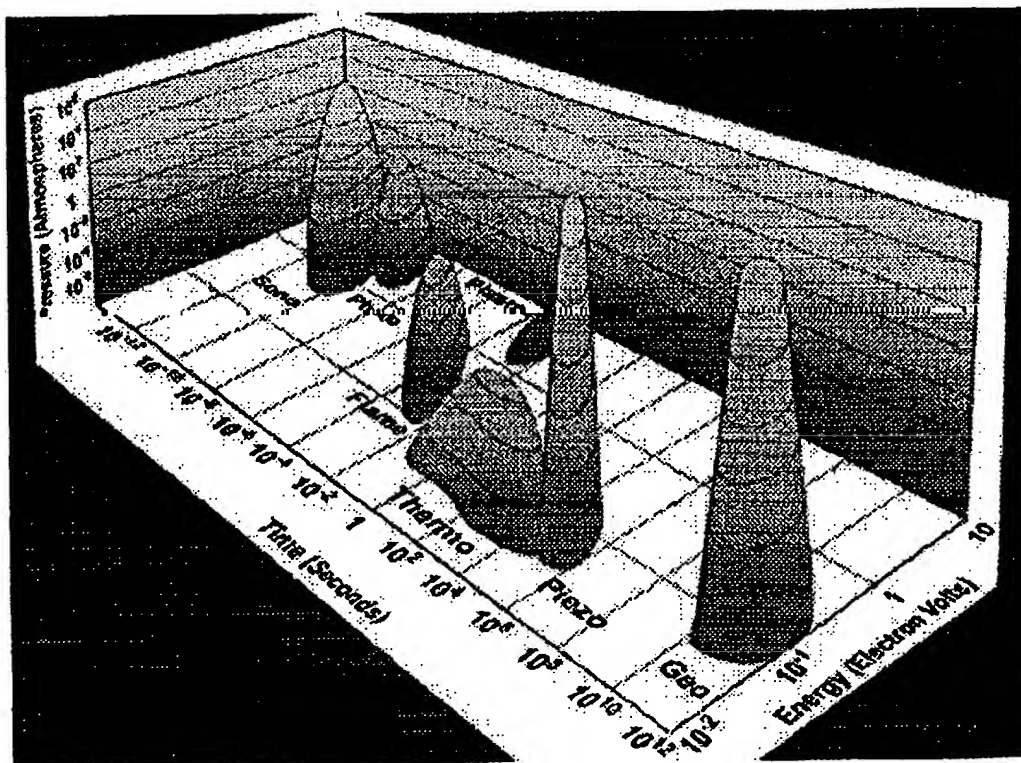


Figure 1. Chemistry: the interaction of energy and matter. The three axes represent duration of the interaction, pressure, and energy per molecule. The labeled islands